



Atty. Dkt. No. 040302-0569

***IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES***

Applicant: Takanori ITOU et al.  
  
Title: POSITIVE ELECTRODE  
MATERIAL FOR NON-  
AQUEOUS ELECTROLYTE  
LITHIUM ION BATTERY AND  
BATTERY USING THE SAME  
  
Appl. No.: 10/581,858  
  
International 11/29/2004  
Filing Date:  
371(c) Date: 06/30/06  
  
Examiner: Jonathan G. LEONG  
  
Art Unit: 1725  
  
Confirmation 4646  
Number:

**BRIEF ON APPEAL**

Mail Stop Appeal Brief - Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

Under the provisions of 37 C.F.R. § 41.37, this Appeal Brief is being filed together with a credit card payment form in the amount of \$540.00 covering the 37 C.F.R. 41.20(b)(2) appeal fee. If this fee is deemed to be insufficient, authorization is hereby given to charge any deficiency (or credit any balance) to the undersigned deposit account 19-0741.

**REAL PARTY IN INTEREST**

The real party in interest is NISSAN MOTOR CO., LTD., a Japanese corporation.

**RELATED APPEALS AND INTERFERENCES**

Appellants are unaware of any appeals or interferences that will directly affect or be directly affected by or have a bearing on the Board's decision in the present appeal.

**STATUS OF CLAIMS**

1. Claims pending: 1-3 and 6
2. Claims cancelled: 4-5 and 7-12
3. Claims withdrawn: None
4. Claims rejected: 1-3 and 6
5. Claims on appeal: 1-3 and 6

A copy of claims 1-3 and 6 is provided in the CLAIMS APPENDIX.

**STATUS OF AMENDMENTS**

In the Amendment and Reply Under 37 CFR § 1.116, filed January 26, 2011, claims 1-3 and 6 were amended and claims 11-12 were cancelled. The amendments were entered in the Advisory Action dated February 15, 2011.

**SUMMARY OF CLAIMED SUBJECT MATTER**

Claim 1 recites a positive electrode material for non-aqueous electrolyte lithium ion battery (see specification at page 5, line 30 to page 6, line 6), comprising: an oxide containing lithium and nickel (see specification at page 5, line 30 to page 6, line 6); and a lithium compound deposited on a surface of the oxide (see specification at page 5, line 30 to page 6, line 6), wherein the lithium compound is lithium sulfate (see specification at page 7, line 14 to page 8, line 2). The claimed subject matter results in an improved “capacitive sustainability rate.”

**GROUND OF REJECTION TO BE REVIEWED ON APPEAL**

Applicants request review of the rejection of claims 1-3 and 6 under 35 U.S.C. § 103(a) over U.S. Patent No. 6,071,649 (“Mao”) in view of U.S. Patent Publication No. 2003/0157490 (“Huang”) as further evidenced by Japanese Patent Publication No. 07-245105 (“Nagayama”). This rejection was initially set forth in the Final Office Action dated October 27, 2010 and was modified by the Advisory Action dated February 15, 2011.

## ARGUMENT

### I. INTRODUCTION

- A. **The present invention is a lithium sulfate deposition layer in a positive electrode for a lithium ion battery, which results in a surprising increase of the “capacitive sustainability rate,” an important lithium ion battery parameter.**

The claimed use of a *lithium nickel base material* and a *lithium sulfate deposition material* in a positive electrode for a lithium ion battery results in a surprising increase of the “capacitive sustainability rate” for the battery.

Positive electrodes for lithium ion batteries come in a wide variety of materials, as is demonstrated in the prior art references cited by the Examiner. Yet the Examiner was unable to cite a single reference that disclosed an electrode that included a *lithium nickel base material* and a *lithium sulfate deposition material*. In fact, as will be discussed in detail below, none of the electrodes in the cited references even suggests such a positive electrode of the present invention. Mr. Takamitsu Saito, one of the co-inventors of the instant invention who discovered the measurable benefits of the present invention, declared:

My sincere belief is that the combination of a lithium nickel oxide and a lithium sulfate being deposited in a positive electrode for a non-aqueous electrolyte lithium ion battery was never known, used, or suggested. ***This is why the present application was filed.***<sup>1</sup>

Because the cited references did not suggest the inventive combination of the present Application, they could not realize the ***entirely unanticipated results*** that the Mr. Saito and his fellow inventors were able to achieve with their invention.

As a brief background, the art of positive electrode materials for non-aqueous lithium ion batteries is today, and was at the time of the invention, heavily influenced by the need to increase battery capacity and, perhaps even more importantly, the need to increase the

---

<sup>1</sup> See Declaration of Takamitsu Saito, Exhibit 1 at paragraph 6; emphasis added.

retention of battery capacity over many uses of cycles for extended periods of time. The quality of a battery can be judged by looking to several battery characteristics, the most important of which is the “*capacity sustainability rate*”.<sup>2</sup>

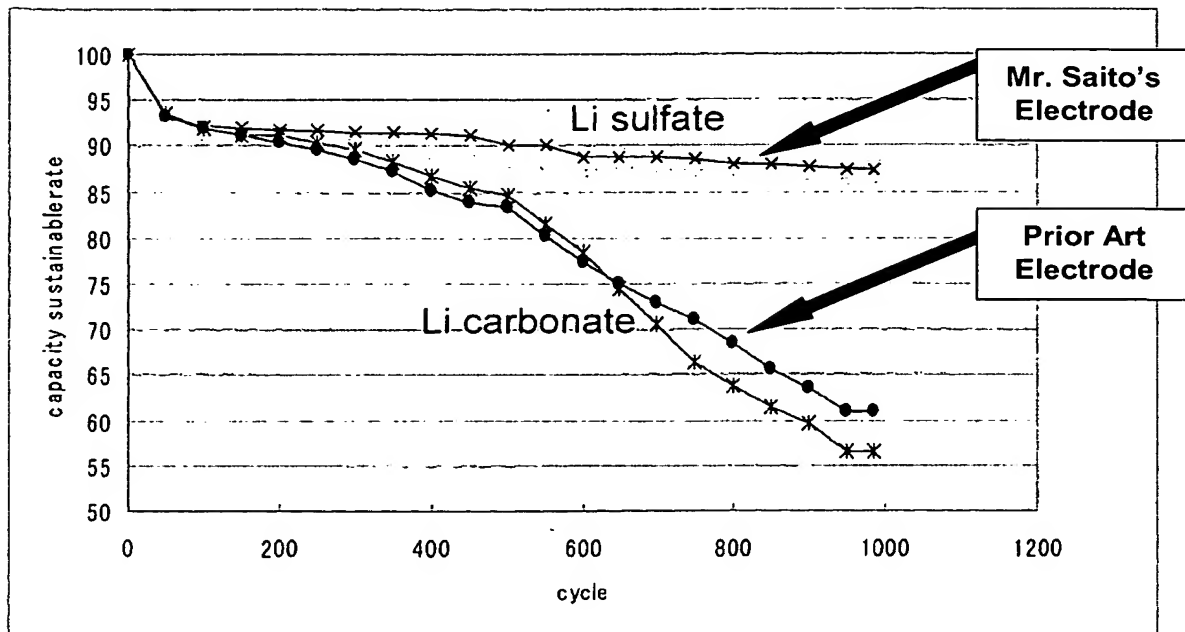
The performance requirements demanded of lithium ion batteries have increased as the power consumption of portable devices has increased due to the enhancement in their performance and functions. The increased performance requirements have led to the use of lithium nickel oxide electrodes, which have a higher capacity than traditional lithium cobalt electrodes.<sup>3</sup> Often, a multitude of laminate-type non-aqueous electrolyte lithium ion batteries connected in series may be used. However, because such batteries are smaller than traditional batteries, the capacity sustainability rate of such laminate-type batteries is particularly important. Unsurprisingly, users of lithium ion batteries want assurance that the batteries will work over many cycles without a substantial decrease in performance—known as the “capacity sustainability rate.”

Mr. Saito and his fellow inventors have been able to improve on the “capacity sustainability rate” through their invention. While the details of the unexpected results provided by their unique electrode will be discussed in II.B below, the following graph below so clearly demonstrates the benefits of the invention as to the “capacity sustainability rate” to be almost self-explanatory:

---

<sup>2</sup> See Specification at page 1, line 23 to page 2, line 11.

<sup>3</sup> See Specification at page 1, lines 23-28.



Note the disturbing downward trend in the prior art lithium carbonate electrodes, representing a substantial decrease in the capacity sustainability rate. In fact the prior art electrodes retain only 60% of their initial capacity after 1000 charge/discharge cycles, while Mr. Saito's electrodes retain almost *90% of their initial capacity after the same number of cycles!* This is why the claimed invention is commercially so important.

These results are obtained through the recited *lithium sulfate deposition layer*. Mr. Saito, an expert in battery design and has extensive experience in the design of non-aqueous electrolyte batteries, has declared that:

***The superiority of lithium sulfate for this use was entirely unexpected.<sup>4</sup>***

Mr. Saito's Declaration and the accompanying experimental results therein provide strong evidence that the claims of the present Application are entirely innovative and non-obvious over the prior art. The Examiner's decision to disregard this evidence is incorrect.

<sup>4</sup>Saito Declaration, Exhibit 1, at paragraph 7.

In fact, the results achieved by the present invention are even more surprising given the complex and generally unpredictable nature of electrode design, which will now be discussed.

**B. The design of positive electrode materials for non-aqueous lithium ion batteries is complex and unpredictable.**

An electrode designer seeking to maximize the performance of a positive electrode is working with a number of constraints. First, the designer must ensure that the results battery has an acceptable swell rate, such that the battery will not rupture at high voltages or temperatures. Swelling is an important parameter for lithium nickel oxide batteries, because in such batteries, oxygen ions are oxidized into oxygen radicals by high valence nickel ions within the positive electrode. The oxygen radicals are released into an electrolyte solution in the battery and cause decomposition of the electrolyte solution. This process generates a large amount of gas when the battery is initially charged or stored at high temperature, which causes the battery to swell.<sup>5</sup> Second, the designer must ensure that battery's capacity remains high enough to satisfy the requirements of the device to be powered by the battery. Third, the designer must ensure that the battery's capacity sustainability rate remains high, such that the battery may be used through multiple cycles without a significant decrease in capacity.

To meet these constraints, the electrode designer has a large number of parameters that can be changed. *First*, he must choose among the potential base materials for the electrode. Two examples of such base materials are discussed above—lithium cobalt oxide and lithium nickel oxide—but many other possibilities exist, including vanadium selenide, titanium disulfide, lithium manganese oxide, lithium iron phosphate, lithium trivanadate, and many others. *Second*, he must choose whether other materials will be included in the electrode. Many electrodes include only a single electrode active material, with no other coatings or additives. The decision to add additional materials is risky, because it can have unexpected effects on the performance of the electrode. *Third*, and most importantly, if additional materials are desired, the electrode designer must choose among the enormous number of potential materials to include in the positive electrode and must also determine

---

<sup>5</sup> See Specification at page 2, line 26 to page 3, line 3.

how much of the additional material to include in the electrode. The instant Specification mentions twenty-two materials<sup>6</sup> that could potentially be used to coat a lithium nickel oxide, but an electrode designer could potentially choose from an extremely large number of materials to include in a positive electrode.

Between any single design parameter (such as electrode material) and the any particular performance criterion (such as capacity sustainability rate) the relationship is rarely one-to-one, such that a parameter can be tweaked and have a predictable and limited impact on the rest of the system. Rather, the design parameters are interrelated, and affect the performance criteria a complex way. For example, adding a lithium compound on the service of a lithium nickel oxide electrode base material might increase the capacity sustainability rate of a battery, but have a detrimental effect on its total capacity.<sup>7</sup>

The evidence thus shows an industry driven to design products that must meet rigorous performance requirements. The performance, however, is measured by criteria that are interrelated functions of numerous design parameters. Furthermore, non-aqueous lithium ion batteries are rarely used under conditions that resemble the assumptions of predictive models, but rather are used in unfavorable environments at extreme temperatures and widely varying humidity.

*The above factors lead to the conclusion that the design of positive electrode materials for non-aqueous lithium ion batteries is complex and unpredictable.*

---

<sup>6</sup> See Specification at Figs. 10-13.

<sup>7</sup> See, e.g., U.S. Patent No. 6,071,649 (“Mao”) at column 4, lines 45-49.

## II. GROUNDS OF REJECTION

### A. The Rejection of claim 1 under 35 U.S.C. § 103(a) should be reversed.

Claim 1 was rejected under 35 U.S.C. § 103(a) as allegedly being unpatentable over U.S. Patent No. 6,071,649 (“Mao”) in view of U.S. Patent Publication No. 2003/0157490 (“Huang”) as evidenced by Japanese Patent Publication No. 07-245105 (“Nagayama”). This rejection should be reversed for at least the following reasons.

1. **The rejection of claim 1 was improper because neither Mau, Huang, nor Nagayama, alone or in combination, discloses, teaches or suggests “a lithium compound deposited on the surface of the oxide, wherein the lithium compound is lithium sulfate.”**

Neither Mau, Huang, nor Nagayama, alone or in combination, discloses, teaches or suggests “a lithium compound deposited on the surface of the oxide, wherein the lithium compound is lithium sulfate,” as required by claim 1. In the Final Office Action, the Examiner acknowledged that Mao does not disclose that the lithium compound is lithium sulfate, but alleged that:

It is well-known in the art that  $\text{LiCoO}_2$  and lithium sulfate are common choices as lithium compounds for use with positive active materials as evidenced by Huang (Claim 5). Thus, it would have been obvious to one having ordinary skill in the art at the time of the invention to use lithium sulfate instead of  $\text{LiCoO}_2$  to cover the oxide containing lithium and nickel as disclosed by Mao since one having ordinary skill in the art at the time of the invention would have had reasonable expectation of success in doing so as evidenced by Huang (Claim 5). Additionally, as long as the lithium compound covers the oxide containing lithium and nickel, one having ordinary skill in the art at the time of the invention would have reasonably expected a success of inhibition of decomposition of the electrolyte solution since the contact surface of the oxide



containing lithium and nickel is decreased (as evidenced by Nagayama [0022]).<sup>8</sup>

Essentially, the Examiner alleged that Mao teaches “a lithium compound deposited on a surface of the oxide,” and that Huang suggests that “the lithium compound is lithium sulfate.” This is incorrect.

Mao teaches a positive electrode active material layer comprising an oxide containing lithium and nickel, and a lithiated transition metal oxide material deposited on the surface of the oxide.<sup>9</sup> In Mao, the lithiated transition metal oxide material is specifically lithium cobalt oxide or cobalt-doped lithium nickel oxide. Mao is quite clear that the lithiated transition metal oxide should contain cobalt, stating that “[c]oating the  $\text{LiNiO}_2$  particles with lithium cobalt oxide or cobalt-doped lithium nickel oxide is preferred since cobalt is less sensitive to moisture than is nickel and since cobalt has a high charge efficiency and stable capacity.”<sup>10</sup> Of course, lithium sulfate, the coating material claimed in claim 1, is *not* a lithiated transition metal oxide material and does *not* contain cobalt, and thus Mao does not disclose, teach, or suggest the combination of features recited in claim 1.

Contrary to the finding of the Examiner, Huang does not remedy the deficiencies of Mao. The Examiner alleged that “it is well-known in the art that  $\text{LiCoO}_2$  and lithium sulfate are common choices as lithium compounds for use with positive active materials as evidenced by Huang (Claim 5).”<sup>11</sup> However, Huang does not, in fact, teach all that the Examiner alleges. Huang teaches a positive electrode that is a composite fabricated by coating a mixture of slurry on an anti-corrosive and high-voltage stable metal foil or grid that acts as a substrate.<sup>12</sup> The slurry includes active materials, polymeric binders, conducting carbon black, and optionally, a catalyst.<sup>13</sup> Huang never suggests that the slurry might include

---

<sup>8</sup> See Final Office Action at page 7.

<sup>9</sup> See Mao at column 2, lines 55-58.

<sup>10</sup> See Mao at column 3, lines 7-10.

<sup>11</sup> See Final Office Action at page 7.

<sup>12</sup> See Huang at paragraph [0022].

<sup>13</sup> *Id.*

an oxide containing lithium and nickel, and lithium sulfate deposited on a surface of the oxide. In fact, Huang never suggests that lithium sulfate be deposited on another material in an electrode. Quite to the contrary, Huang teaches that the *positive electrode active material itself* can be selected from an expansive list of materials that happens to include both lithium cobalt oxide and lithium sulfate. The active material corresponds to the “oxide containing lithium and nickel” of claim 1, not the lithium compound deposited on the surface of the oxide. A summary of the relevant teachings of Mao, Huang, and Nagayama is presented in the chart below:

Reference	Active (Base) Material	Compound Deposited on Active Material
Mao	Lithium Nickel Oxide	Lithium Cobalt Oxide
Huang	Lithium Sulfate (among many others)	None
Nagayama	Lithium Nickel Oxide	Lithium Carbonate

Nowhere does Huang teach that a lithium compound (much less lithium sulfate) is deposited on the surface of the oxide, as required by claim 1 of the present application. In fact, lithium sulfate is not even discussed anywhere in the Huang specification other than claim 5.

Nor is lithium sulfate mentioned anywhere in Mao or Nagayama. For at least these reasons, claim 1 is allowable over Mao and Huang.

**2. The rejection of claim 1 was improper because the prior art provides no motivation to combine Mao, Huang, and Nagayama.**

Even if the combination of prior art references were to disclose the features of claim 1 (which they do not), the rejection of claim 1 was improper because the prior art provides no motivation to combine Mao, Huang, and Nagayama. The Supreme Court in *KSR Int'l Co. v. Teleflex, Inc.*,<sup>14</sup> when clarifying the standards for obviousness, stated that “a patent composed of several elements is not proved obvious merely by demonstrating that each of its elements was, independently, known in the art . . . . [I]t can be important to *identify a reason* that

---

<sup>14</sup> 550 U.S. 398 (2007)

would have prompted a person of ordinary skill in the relevant field to combine the elements in the way the claimed new invention does.”<sup>15</sup>

The Examiner did not allege that either Mao, Huang, or Nagayama provide an explicit motivation to combine the references. Instead, the Examiner alleged that:

[I]t would have been obvious to one having ordinary skill in the art at the time of the invention to use lithium sulfate instead of  $\text{LiCoO}_2$  to cover the oxide containing lithium and nickel as disclosed by Mao since one having ordinary skill in the art at the time of the invention would have had reasonable expectation of success in doing so as evidenced by Huang (Claim 5). Additionally, as long as the lithium compound covers the oxide containing lithium and nickel, one having ordinary skill in the art at the time of the invention would have reasonably expected a success of inhibition of decomposition of the electrolyte solution since the contact surface of the oxide containing lithium and nickel is decreased (as evidenced by Nagayama [0022])<sup>16</sup>

A proper application of the Office Guidelines for evaluating obviousness under *KSR v. Teleflex* compels the conclusion that claim 1 is patentable over Mao, Huang, and Nagayama in any combination.

The Office Guidelines under KSR lay out several structured approaches (or “rationales”) for evaluating obviousness. The Office Guidelines provide several elements of required proof for establishing a prima facie case of obviousness under each rationale. The appropriate rationale is uncertain here, because the Examiner does not explicitly apply the Office Guidelines. However, given the Examiner’s repeated use of the phrase “reasonable expectation of success,” the rationale that seems closest to the Examiner’s line of reasoning is

---

<sup>15</sup> *KSR*, 550 U.S. at 401.

<sup>16</sup> See Final Office Action at page 7.

rationale E, which is entitled “‘Obvious To Try’ - Choosing From a Finite Number of Identified, Predictable Solutions, With a Reasonable Expectation of Success.” The Office Guidelines state for this rationale that:

To reject a claim based on this rationale, Office personnel must resolve the Graham factual inquiries. Then, Office personnel must articulate the following: (1) a finding that at the time of the invention, there had been a recognized problem or need in the art, which may include a design need or market pressure to solve a problem; (2) a finding that there had been a finite number of identified, predictable potential solutions to the recognized need or problem; (3) a finding that one of ordinary skill in the art could have pursued the known potential solutions with a reasonable expectation of success; and (4) whatever additional findings based on the Graham factual inquiries may be necessary, in view of the facts of the case under consideration, to explain a conclusion of obviousness.

Office Guidelines, p. 57532; MPEP 2143(E).

Importantly and correctly, the Office guidelines stress that “*[i]f any of these findings cannot be made, then this rationale cannot be used to support a conclusion that the claim would have been obvious to one of ordinary skill in the art.*” Office Guidelines, p. 57532 (emphasis added); MPEP 2143(E).

Thus, the Office Guidelines under KSR require proof of each of the following elements to establish a *prima facie* case of obviousness under rationale E:

- (1) The cited references must have recognized a problem or need in the art.
- (2) There must have been only a finite number of solutions to the recognized problem.
- (3) The results of the solutions must have been predictable and likely to be successful.

In the present case, the Final Office Action failed to establish that there was a recognized problem with a finite number of solutions. Furthermore, the Final Office Action failed to establish that the results of the solutions were predictable and likely to be successful.

**a. The Final Office Action failed to establish that there was a recognized problem with a finite number of solutions.**

The Final Office Action failed to establish that there was a recognized problem with a finite number of solutions. Courts have made clear that, to find that a claim is obvious under rationale E, the “finite number” of solutions must be quite small. For example, in *Perfect Web v. Infousa*,<sup>17</sup> the Federal Circuit held that a claim to a method of managing bulk email distribution was obvious where there were only *three potential solutions* to a recognized problem.<sup>18</sup> In *Bayer Schering Pharma AG v. Barr Lab.*,<sup>19</sup> the Federal Circuit held that a pharmaceutical composition was obvious where there were only *two potential solutions* to a recognized problem.<sup>20</sup> The present Application is entirely distinguishable from the cases in which a “finite number of potential solutions” was found.

As discussed in Section I.B above, there are an unfathomably large number of materials that could possibly be deposited on the surface of the oxide of claim 1.

The Examiner alleged that “[I]t would have been obvious to one having ordinary skill in the art at the time of the invention to use lithium sulfate instead of  $\text{LiCoO}_2$  to cover the oxide containing lithium and nickel as disclosed by Mao since one having ordinary skill in the art at the time of the invention would have had reasonable expectation of success in doing so as evidenced by Huang (Claim 5).”<sup>21</sup> As discussed in Section II.A.1 above, Huang does not suggest that any material be deposited on a lithium nickel oxide, and thus the materials in Huang are not “identified, predictable solutions” to the problem recognized in Nagayama. But even in the unlikely event that a person of ordinary skill would be somehow be motivated

---

<sup>17</sup> 587 F.3d 1324 (Fed. Cir. 2009).

<sup>18</sup> *Id.* at 1331.

<sup>19</sup> 575 F.3d 1341 (Fed. Cir. 2009).

<sup>20</sup> *Id.* at 1350.

<sup>21</sup> *Id.*

to choose a *deposition material* from the *active materials* disclosed in Huang, the potential number of materials is enormous. Huang discloses that “the active material of the positive electrode element is selected from the group consisting of lithium intercalation compounds, lithium salts, lithium oxides.”<sup>22</sup> Even the specific examples of these compounds given in Huang is quite large, including over twenty-six different materials.<sup>23</sup> To find that the large number of materials disclosed in Huang are “finite number of identified, predictable potential solutions” would fly in the face of the Federal Circuit’s precedent regarding the correct use of the “obvious to try” rationale.

The Examiner further alleged that “as long as the lithium compound covers the oxide containing lithium and nickel, one having ordinary skill in the art at the time of the invention would have reasonably expected a success of inhibition of decomposition of the electrolyte solution since the contact surface of the oxide containing lithium and nickel is decreased (as evidenced by Nagayama [0022]).”<sup>24</sup> Yet Nagayama suggests only that lithium carbonate can be deposited on the surface of the oxide, and does not suggest the use of any other materials, much less lithium sulfate. To the extent that the Examiner believed that Nagayama suggests that *any material* could be deposited on a lithium nickel oxide, Nagayama suggests no such thing. And even if it did, this would simply support the Applicants’ contention that there were not, in fact, a “finite number of identified, predictable potential solutions” to a problem recognized in the art.

**b. The Final Office Action failed to establish that the results of the solutions were predictable and likely to be successful.**

The Final Office Action failed to establish that the results of the solutions were predictable and likely to be successful. As discussed in Section I.B above, the design of positive electrode materials for non-aqueous lithium ion batteries is complex and unpredictable.

---

<sup>22</sup> See Huang at claim 5.

<sup>23</sup> *Id.*

<sup>24</sup> See Final Office Action at page 7.

As discussed above, Huang does not teach that a lithium compound (much less lithium sulfate) is deposited on the surface of the oxide. So why would a person of ordinary skill believe that such a deposited lithium compound would yield a successful positive electrode material? The simple answer is that he would not! The only actual testing of a lithium nickel oxide having a lithium compound deposited on its surface are in Mao and Nagayama, but these tests were performed only for lithium cobalt oxide and lithium carbonate. Nowhere do the prior art references disclose, teach or suggest that positive electrodes with other deposition materials might yield successful results. Furthermore, the level of success achieved by Nagayama pales in comparison to the success achieved by the present invention, as will next be discussed.

**3. The rejection of claim 1 was improper because the unexpected results achieved by the invention embodying claim 1 demonstrate that claim 1 is not obvious over Mao, Huang, and Nagayama, either alone or in any combination.**

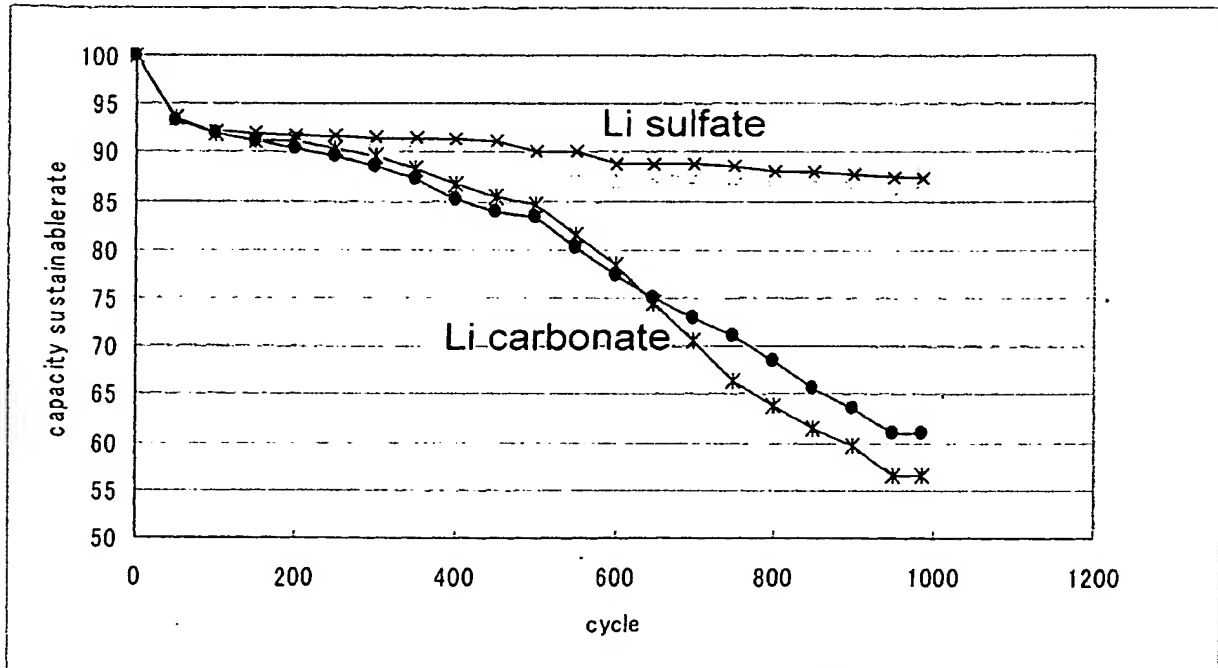
The rejection of claim 1 was improper because the unexpected results achieved by the invention embodying claim 1 demonstrate that claim 1 is not obvious over Mao, Huang, and Nagayama, either alone or in any combination. The MPEP provides that “[e]vidence of unobvious or unexpected advantageous properties, such as superiority in a property the claimed compound shares with the prior art, can rebut prima facie obviousness.”<sup>25</sup> The Examiner acknowledged, but summarily dismissed Applicants’ evidence of unexpected results provided in the Declaration of Takamitsu Saito, filed January 26, 2011 (“Saito Declaration”). This was a clear error.

The Saito Declaration, attached as Exhibit 1, provides strong evidence that the invention embodying claim 1 yields entirely unexpected results. Specifically, the Saito Declaration and experimental results shown therein demonstrate that an electrode material comprising an oxide containing lithium and nickel with lithium sulfate deposited on the oxide yields a surprisingly excellent “capacity sustainability rate,” which, as discussed in Section I.A above, is an important metric in the design of positive electrodes for non-aqueous lithium ion batteries. The results of experiments comparing lithium carbonate and lithium sulfate

---

<sup>25</sup> MPEP § 716.02(a) (citing *In re Chupp*, 816 F.2d 643, 646 (Fed. Cir. 1987)).

deposition materials is shown in the graph on page 2 of the Saito Declaration, reproduced below:



The graph demonstrates that a battery using the electrode material of claim 1 retains nearly 90% of its initial capacity after 1000 cycles, while a battery using an electrode material with lithium carbonate instead of lithium sulfate retains, at best, slightly over 60% of its initial capacity after 1000 cycles. In the Saito Declaration, Mr. Takamitsu Saito, a co-inventor with extensive experience in the design of non-aqueous electrolyte batteries, states that “[t]he superiority of lithium sulfate for this use was entirely unexpected.” See Saito Declaration at paragraph 5-7; see also Amendment and Reply Under 37 C.F.R. § 1.116 filed January 26, 2011 at pages 5-7. Yet, citing various informalities in the evidence, the Patent Office concluded that it “cannot determine if there are unexpected results . . . .”<sup>26</sup>

The Federal Circuit has held that “when an applicant demonstrates substantially improved results . . . and states that the results were unexpected, this should suffice to establish unexpected results in the absence of evidence to the contrary.” *In re Soni*, 54 F.3d

<sup>26</sup> See Advisory Action mailed February 15, 2011 at Continuation Sheet.



746, 751 (Fed. Cir. 1995) (also holding that “[g]iven the presumption of similar properties for similar compositions, substantially improved properties are ipso facto unexpected.”).

Applicants have demonstrated substantially improved results and stated, in the Saito Declaration, that the results were unexpected, thus satisfying the criteria of *In re Soni*. This suffices to establish unexpected results in the absence of evidence to the contrary.

The Office also alleges that “the evidence of unexpected results is deficient at least since it is not reasonably commensurate in scope with the prior art . . . because the evidence has a coating of lithium carbonate while Mao discloses a coating of lithium cobaltate.”<sup>27</sup> The MPEP states that “[a]n affidavit or declaration under 37 CFR 1.132 must compare the claimed subject matter with the *closest prior art* to be effective to rebut a prima facie case of obviousness.”<sup>28</sup> At the time the Saito Declaration was filed, the closest references cited by the Patent Office were Nagayama and U.S. Patent No. 5,427,875 (“Yamamoto”), both of which teach an electrode material comprising lithium nitride coated with lithium carbonate. This is why evidence was presented comparing lithium sulfate and lithium carbonate. This is all that is required by the MPEP, and thus the Office’s failure to give adequate weight to the Saito Declaration was a clear error.

For at least these reasons, claim 1 is patentable over Mao, Huang, and Nagayama, alone or in any combination.

**B. The Rejection of claims 2, 3, and 6 under 35 U.S.C. § 103(a) should be reversed.**

Claims 2, 3, and 6 depend from independent claim 1, and are allowable for at least the same reasons as claim 1, even without regard to the further patentable features recited therein.

---

<sup>27</sup> See Advisory Action mailed February 15, 2011 at Continuation Sheet.

<sup>28</sup> MPEP § 716.02(e) (citing *In re Burckel*, 592 F.2d 1175, 201 USPQ 67 (CCPA 1979)) (emphasis added).

**III. CONCLUSION**

In view of the foregoing, it is respectfully requested that the Board reverse the rejections of claims 1-3 and 6.

Respectfully submitted,

Date 8/9/2011

By Michael D. Kaminski

FOLEY & LARDNER LLP  
Customer Number: 22428  
Telephone: (202) 672-5490  
Facsimile: (202) 672-5399

Michael D. Kaminski  
Attorney for Applicants  
Registration No. 32,904

Chase J. Brill  
Attorney for Applicants  
Registration No. 61,378

**CLAIMS APPENDIX**

1. (Previously Presented) A positive electrode material for non-aqueous electrolyte lithium ion battery, comprising:
  - an oxide containing lithium and nickel; and
  - a lithium compound deposited on a surface of the oxide, wherein the lithium compound is lithium sulfate.
2. (Previously Presented) A positive electrode material according to claim 1, wherein the lithium compound is deposited to cover substantially an entire surface of the oxide, and a thickness of a cover layer of the lithium compound ranges from 5 nm to 1  $\mu$ m.
3. (Previously Presented) A positive electrode material according to claim 1, wherein the lithium compound is deposited to sprinkle on the surface of the oxide, and a volume of the lithium compound ranges from 0.5 to 10 % with respect to a volume of the positive electrode active material.
- 4-5. (Canceled)
6. (Previously Presented) A non-aqueous electrolyte lithium ion battery, comprising:
  - a positive electrode active material layer comprising a positive electrode material according to claim 1;
  - a negative electrode active material layer comprising a negative electrode active material; and
  - an electrolyte layer disposed between the positive and negative electrode active materials layers.
- 7-12. (Cancelled)

**EVIDENCE APPENDIX**

1. Declaration Under 37 C.F.R. § 1.132, filed with Applicants' Response dated January 26, 2011, and entered by the Examiner in the Advisory Action dated February 14, 2011.
2. Japanese Patent Publication No. 07-245105 ("Nagayama"), filed with Applicants' Information Disclosure Statement dated September 23, 2010, and entered by the Examiner in the Final Rejection dated October 27, 2010.

The remaining references cited by the Examiner in establishing the grounds of rejection in the Final Office Action dated October 27, 2010 are all U.S. Patent documents. Accordingly, copies of these references have not been enclosed.

**RELATED PROCEEDINGS APPENDIX**

None.

## **Evidence Exhibit 2**

# EUROPEAN PATENT OFFICE

Ed

## Patent Abstracts of Japan

PUBLICATION NUMBER : 07245105  
PUBLICATION DATE : 19-09-95

APPLICATION DATE : 04-03-94  
APPLICATION NUMBER : 06034366

APPLICANT : MATSUSHITA ELECTRIC IND CO LTD;

INVENTOR : NAGAYAMA MASATOSHI;

INT.CL. : H01M 4/58 H01M 4/02 H01M 10/40

TITLE : NONAQUEOUS ELECTROLYTE SECONDARY BATTERY AND POSITIVE ELECTRODE ACTIVE MATERIAL THEREOF

ABSTRACT : PURPOSE: To provide a battery having excellent discharge performance after it is preserved at a high temperature by improving a positive electrode active material in a secondary battery using nonaqueous electrolyte.

CONSTITUTION: A positive electrode active material where the whole surface or a part of a surface of nickel acid lithium is covered with lithium carbonate is used. Thereby, a contact surface of the nickel acid lithium and nonaqueous electrolyte is reduced, and formation of a film which is thought to be created when the electrolyte is decomposed and checks discharge reaction is eliminated, and the degradation of discharge performance after a battery is preserved at a high temperature can be improved.

COPYRIGHT: (C)1995,JPO

(19) 日本国特許庁 (J P)

(12) 公開特許公報 (A)

(11) 特許出願公開番号

特開平7-245105

(43) 公開日 平成7年(1995)9月19日

(51) Int.Cl. <sup>4</sup>	識別記号	庁内整理番号	F I	技術表示箇所
H 0 1 M	4/58			
	4/02	B		
	10/40	Z		

審査請求 未請求 請求項の数 2 O L (全 4 頁)

(21) 出願番号 特願平6-34366

(22) 出願日 平成6年(1994)3月4日

(71) 出願人 000005821

松下電器産業株式会社  
大阪府門真市大字門真1006番地

(72) 発明者 小林 茂雄

大阪府門真市大字門真1006番地 松下電器  
産業株式会社内

(72) 発明者 山浦 純一

大阪府門真市大字門真1006番地 松下電器  
産業株式会社内

(72) 発明者 岡村 一広

大阪府門真市大字門真1006番地 松下電器  
産業株式会社内

(74) 代理人 弁理士 小銀治 明 (外2名)

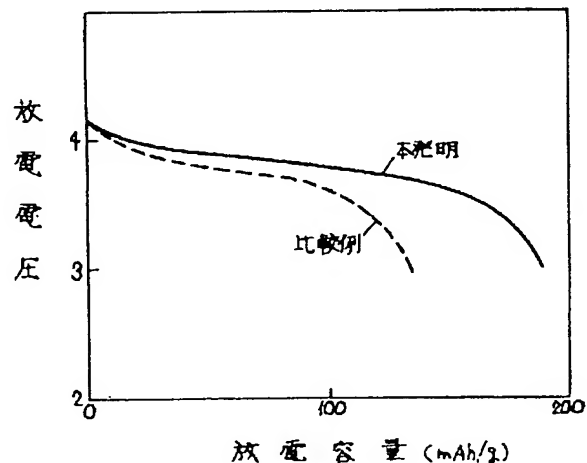
最終頁に続く

(54) 【発明の名称】 非水電解液二次電池とその正極活物質

(57) 【要約】

【目的】 非水電解液を用いた二次電池であり、正極活物質を改善することで、高温保存後の放電性能に優れた電池を提供する。

【構成】 ニッケル酸リチウムの表面の全面もしくは一部が炭酸リチウムで被覆されている正極活物質を用いた。これによりニッケル酸リチウムと非水電解液との接触面を減少させ、電解液が分解してできるとされる放電反応を阻害する膜の形成をなくし、高温保存後における放電性能の劣化を改善することができる。





## 【特許請求の範囲】

【請求項1】 ニッケル酸リチウムの表面の全面もしくは一部が炭酸リチウムで被覆されている非水電解液二次電池の正極活物質。

【請求項2】 表面の全面もしくは一部が炭酸リチウムで被覆されているニッケル酸リチウムを正極活物質、リチウムを負極活物質とし、非水電解液を備えた非水電解液二次電池。

## 【発明の詳細な説明】

【0001】

【産業上の利用分野】本発明は、非水電解液二次電池、特にリチウム複合酸化物を正極活物質材料に用いた電池の改良に関するものである。

【0002】

【従来の技術】近年、AV機器あるいはパソコン等の電子機器のポータブル化、コードレス化が急速に進んでおり、これらの駆動用電源として小型、軽量で高エネルギー密度を有する二次電池への要求が高い。このような点で非水系二次電池、特にリチウム二次電池は、とりわけ高電圧、高エネルギー密度を有する電池として期待が高い。

【0003】上記の要望を満たす正極活物質としてリチウムをインタカレートすることのできるニッケル酸リチウム ( $\text{LiNiO}_2$ ) が提案されている。しかしながら高温に放置すると活物質の劣化が大きく、放電性能が悪くなるなどの課題があり、未だ商品化には至っていない。

【0004】一方、 $\text{LiNiO}_2$ は炭酸ガスと反応して炭酸リチウムと酸化ニッケルになることが知られている (Journal of Catalyst 132, 92, 1991) が、炭酸リチウムで被覆された  $\text{LiNiO}_2$  が電池の正極活物質として有効かどうかは知られていない。

【0005】

【発明が解決しようとする課題】 $\text{LiNiO}_2$ は、リチウムに対して4V以上の電位を示し、これを正極活物質として用いると高エネルギー密度を有する二次電池が期待できる。しかしその保存特性は悪く、とくに高温での保存における容量劣化率は大きい。

【0006】

【課題を解決するための手段】上記の課題を解決するため本発明は、正極活物質として表面の全面もしくは一部が炭酸リチウムで被覆された  $\text{LiNiO}_2$  を用いるものである。このような正極活物質材料を用いることにより、保存特性、特に高温保存における容量劣化率の小さい非水電解液二次電池が得られることを見だしたものである。

【0007】

【作用】 $\text{LiNiO}_2$ は高温保存後における放電容量の劣化率が大きい。その原因は定かでないが、 $\text{LiNiO}_2$

と非水電解液との直接接触は非水電解液を分解し、 $\text{LiNiO}_2$ の表面に放電反応を阻害する被膜が形成される。これが良好な高温保存における放電容量の劣化を大きくする一つの因子と考えられる。

【0008】本発明では、正極活物質である  $\text{LiNiO}_2$  の表面の全面もしくは一部を炭酸リチウムで被覆することにより、 $\text{LiNiO}_2$ と非水電解液との接触面を減少させ、電解液が分解してできると思われる電解液との反応膜の形成を無くし、高温保存後の放電性能の劣化を改善しようとするものである。

【0009】

【実施例】以下、本発明を図とともに具体的な実施例に沿って説明する。

【0010】表面が炭酸リチウムで被覆された正極活物質  $\text{LiNiO}_2$  の作成方法は、水酸化リチウムと水酸化ニッケルをリチウムとニッケルとが化学量論で1:1になるよう配合し、混合の後700℃で5時間焼成する。その際の焼成雰囲気は酸素雰囲気とした。次に炭酸リチウムで  $\text{LiNiO}_2$  の表面を被覆する。炭酸リチウムで表面を被覆する方法としては、炭酸ガス雰囲気中に150℃で2~3分放置するか、もしくは空气中に200℃で10分放置することにより、 $\text{LiNiO}_2$ の表面を炭酸リチウムで被覆する。

【0011】図1にそのモデル図を示す。炭酸リチウムの確認はX線光電子分光分析もしくは赤外分光分析によって確認できる。また被覆状態は  $\text{LiNiO}_2$  表面に層状に密着しており、その層の厚みは10~100

【0012】

【外1】

A

【0013】程度である。次に得られた正極活物質を電池に用いた際の高温保存における放電容量劣化率を評価するため円筒型電池を試作して検討した。上記の正極活物質100重量部、アセチレンブラック4重量部、フッ素樹脂系結着剤7重量部を混合して正極合剤とし、カルボキシメチルセルロース水溶液に懸濁させてペースト状にした。このペーストをアルミニウム箔の両面に塗着し、乾燥後圧延して極板とした。

【0014】負極は、コークスを焼成して粉砕した炭素材100重量部に、フッ素樹脂系結着剤10重量部を混合し、カルボキシメチルセルロース水溶液に懸濁させてペースト状にした。そしてこのペーストを銅箔の両面に塗着し、乾燥後圧延して負極板とした。

【0015】図2にこれらの極板を用いて構成した円筒形電池の縦断面図を示す。電池の構成は帯状の正極板、負極板それぞれにリードを取りつけ、ポリプロピレン製のセパレータを介して渦巻き状に巻回し、これを電池ケース内に収納した。電解液にはプロピレンカーボネートとエチレンカーボネートとの等容積混合溶媒に、過塩素酸リチウムを1モル/1の割合で溶解したものをを用い、

これを所定量注入してケースを封口したものを試験電池とした。

【0016】この図2において、1は耐有機電解液性のステンレス銅板を加工した電池ケース、2は安全弁を設けた封口板、3は絶縁パッキングを示す、4は極板群であり、正極板および負極板がセパレータを介して渦巻き状に巻回されてケース内に収納され、正極リード5は封口板2に接続され、負極からは負極リード6が引き出されて電池ケース1の底部に接続されている。7は絶縁リングで極板群4の上下部にそれぞれ設けられている。

【0017】この試験電池を充放電電流100mA、充電終止電圧4.1V、放電終止電圧3.0Vの条件下で定電流充放電試験を5サイクルまで常温で行い、その後60℃で20日間放置し、上記と同様の充放電試験を常温で行った。

【0018】図3に60℃で20日間放置後の放電性能を示す。比較例として従来の炭酸リチウムが表面に被覆されていない $\text{LiNiO}_2$ の放電特性もあわせて示した。

【0019】図3からわかるように、炭酸リチウムで表面が被覆されている $\text{LiNiO}_2$ は60℃で20日間放置後も優れた放電性能を示すが、表面が何ら被覆されていない $\text{LiNiO}_2$ は放電容量が劣化している。

【0020】本実施例では負極として、リチウムが吸着もしくはインタカレートする炭素材を用いたが、この他にリチウム金属、リチウム合金を用いても同様な効果が

得られる。なおニッケルの一部を他の遷移金属に置き換えてもほぼ同様の結果が得られた。

【0021】また炭酸リチウムの被膜は $\text{LiNiO}_2$ の表面全体を完全に覆うことなく、その一部分のみを被覆している状態であっても同様な結果が得られた。

【0022】

【発明の効果】以上から明らかなように、本発明によれば正極活物質として、表面が炭酸リチウムで被覆された $\text{LiNiO}_2$ を用いることにより、 $\text{LiNiO}_2$ と非水電解液との接触面を減少させ、電解液が分解してできるとされる放電反応を阻害する膜の形成を無くすことができ、高温保存後においても放電性能に優れた非水電解液二次電池を得ることができる。

【図面の簡単な説明】

【図1】本発明を説明するモデル図

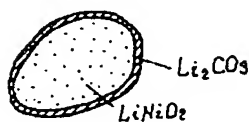
【図2】本発明の実施例における円筒形電池の縦断面図

【図3】本発明と比較例電池の放電性能の比較を示す図

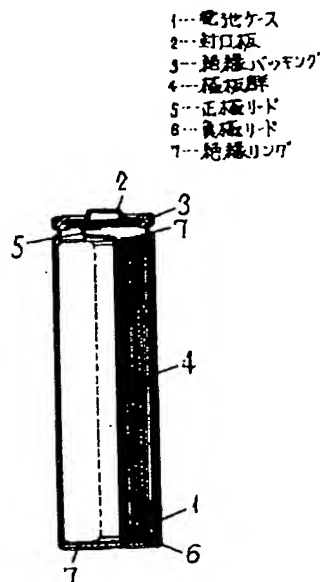
【符号の説明】

- 1 電池ケース
- 2 封口板
- 3 絶縁パッキング
- 4 極板群
- 5 正極リード
- 6 負極リード
- 7 絶縁リング

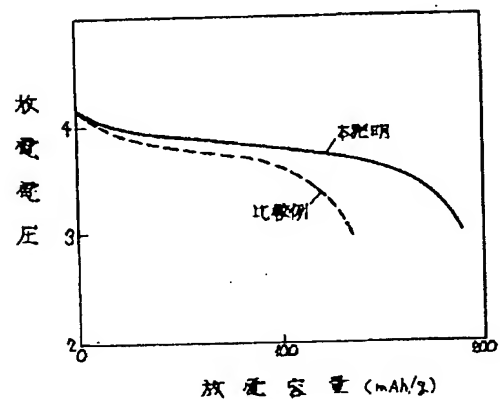
【図1】



【図2】



【図3】



フロントページの続き

(72)発明者 井上 薫  
大阪府門真市大字門真1006番地 松下電器  
産業株式会社内

(72)発明者 永山 雅敏  
大阪府門真市大字門真1006番地 松下電器  
産業株式会社内

**Disclaimer:**

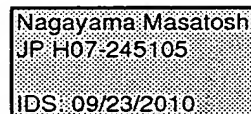
This English translation is produced by machine translation and may contain errors. The JPO, the INPIT, and those who drafted this document in the original language are not responsible for the result of the translation.

**Notes:**

1. Untranslatable words are replaced with asterisks (\*\*\*\*).
2. Texts in the figures are not translated and shown as it is.

Translated: 02:30:27 JST 10/13/2010

Dictionary: Last updated 10/08/2010 / Priority:



---

**FULL CONTENTS**

---

**[Claim(s)]**

[Claim 1]The quality of cathode active material of a nonaqueous electrolyte rechargeable battery with which the whole surface or some of surface nickel acid lithium is covered by lithium carbonate.

[Claim 2]A nonaqueous electrolyte rechargeable battery which surface [ the whole surface or some of ] made nickel acid lithium covered with lithium carbonate quality of cathode active material, made lithium an anode active material, and was provided with nonaqueous electrolyte.

---

**[Detailed Description of the Invention]****[0001]**

[Industrial Application]This invention relates to improvement of a nonaqueous electrolyte rechargeable battery, especially the battery which used the lithium compound oxide for the nature material of cathode active material.

**[0002]**

[Description of the Prior Art]In recent years, portable-izing of electronic equipment, such as AV equipment or a personal computer, and cordless-ization are progressing quickly, and the demand to small size and the rechargeable battery which is lightweight and has high energy density is high as these power supplies for a drive. A nonaqueous system rechargeable battery, especially a lithium secondary battery have high expectation at such a point as a battery which especially has a high voltage and high energy density.

[0003]The nickel acid lithium ( $\text{LiNiO}_2$ ) which can carry out the in hawk rate of the lithium as quality of cathode active material which fills the above-mentioned request is proposed.

However, when it is neglected to high temperature, degradation of an active material is large, SUBJECT, like electric discharge performance worsens occurs, and it has not yet resulted in commercialization.

[0004]on the other hand,  $\text{LiNiO}_2$  reacting to carbon dioxide and becoming lithium carbonate and nickel oxide is known -- \*\*\*\* (Journal of Catalyst 132, 92, and 1991). It is not known whether  $\text{LiNiO}_2$  covered with lithium carbonate is effective as the quality of cathode active material of a battery.

[0005]

[Problem to be solved by the invention]If  $\text{LiNiO}_2$  shows the potential beyond 4V to lithium and this is used as quality of cathode active material, it can expect the rechargeable battery which has high energy density. However, the preservation characteristic is bad and especially the capacity degradation rate in the preservation in high temperature is large.

[0006]

[Means for solving problem]In order to solve above-mentioned SUBJECT,  $\text{LiNiO}_2$  by which surface [ the whole surface or some of ] was covered by lithium carbonate is used for this invention as quality of cathode active material. By using such a nature material of cathode active material, it is begun to see a nonaqueous electrolyte rechargeable battery with small preservation characteristic, especially capacity degradation rate in high temperature preservation obtained.

[0007]

[Function] $\text{LiNiO}_2$  has a large degradation rate of the electric discharge capacity after high temperature preservation. Although the cause is not certain, the contact with  $\text{LiNiO}_2$  and nonaqueous electrolyte decomposes nonaqueous electrolyte directly, and the film which checks an electric discharge reaction is formed in the surface of  $\text{LiNiO}_2$ . This is considered to be one factor which enlarges degradation of the electric discharge capacity in good high temperature preservation.

[0008][ by lithium carbonate covering the whole surface or a part of surface  $\text{LiNiO}_2$  which is quality of cathode active material in this invention ] The contact surface of  $\text{LiNiO}_2$  and nonaqueous electrolyte tends to be decreased, formation of a reaction film with the electrolysis solution considered that an electrolysis solution is decomposed and made tends to be lost, and it is going to improve degradation of the electric discharge performance after high temperature preservation.

[0009]

[Working example]Hereafter, this invention is explained in accordance with a concrete

embodiment with a figure.

[0010]The preparation method of nature  $\text{LiNiO}_2$  cathode active material by which the surface was covered with lithium carbonate blends lithium hydroxide and nickel hydroxide so that lithium and nickel may be set to 1:1 by chemicals stoichiometry, and it calcinates them at 700 °C after mixture for 5 hours. Calcination atmosphere in that case was made into oxygen environment. Next, the surface of  $\text{LiNiO}_2$  is covered with lithium carbonate. The surface of  $\text{LiNiO}_2$  is covered with lithium carbonate by neglecting it at 150 °C in carbon dioxide atmosphere for 2 to 3 minutes, or neglecting it at 200 °C in the air as a method of covering the surface with lithium carbonate, for 10 minutes.

[0011]The model figure is shown in drawing 1. The check of lithium carbonate can be checked by X ray photoelectron spectrometry or infrared emission spectroscopy. The covering state is stuck to the  $\text{LiNiO}_2$  surface in layers, and the thickness of the layer is 10-100 nm. [0012]

[External character 1]

Å

[0013]It is a grade. Next, the cylindrical battery was made as an experiment and examined in order to evaluate electric discharge capacity degradation in the high temperature preservation at the time of using the obtained quality of cathode active material for a battery. Mixed the nature of cathode active material 100 above-mentioned weight section, acetylene black 4 weight section, and fluoro-resin system binder 7 weight section, considered it as the cathode medical mixture, carboxymethyl cellulose solution was made suspended, and it was made the shape of a paste. Both sides of aluminum foil were plastered with this paste, and it rolled after dryness, and was considered as the polar plate.

[0014]The anode mixed fluoro-resin system binder 10 weight section to carbon material 100 weight sections which calcinated and ground corks, made carboxymethyl cellulose solution suspended to them, and was made into the shape of a paste at them. And both sides of copper foil were plastered with this paste, and it rolled after dryness, and was considered as the anode board.

[0015]The longitudinal section of the cylindrical shape battery which used and constituted these polar plates is shown in drawing 2. The composition of the battery attached the lead to a band-like cathode board and each anode board, and stored winding and this in the cell case spirally via the separator made from polypropylene POREN. What carried out specified quantity pouring of this, and obturated the case was made capacity mixed solvents, such as propylene carbonate and ethylene carbonate, with the examination battery at the electrolysis solution using what dissolved lithium perchlorate at a rate of 1 mol / l.

[0016]this drawing 2 \*\*\*\* -- the cell case into which 1 processed the stainless steel plate of

organic electrolysis-proof liquefaction, the obturation board with which 2 provided the safety valve, and 3 show insulating packing. A cathode board and an anode board are spirally wound via a separator, and it is stored in a case, and from the anode, 4 is plates very much and the cathode lead 5 is connected [ it is connected to the obturation board 2, the anode lead 6 is pulled out, and ] to the bottom of the cell case 1. 7 is provided very much in the vertical section of the plates 4 with the insulating ring, respectively.

[0017]The constant-current charge test was done at normal temperature up to 5 cycles under the conditions of 100 mA of charge and discharge currents, the charge final voltage 4.1V, and the electric discharge final voltage 3.0V, this examination battery was neglected for 20 days at 60 °C after that, and the same charge and discharge test as the above was done at normal temperature.

[0018]60 °C shows the electric discharge performance after neglect to drawing 3 for 20 days. The electric discharge characteristic of  $\text{LiNiO}_2$  with which conventional lithium carbonate is not covered by the surface as a comparative example was also united and shown.

[0019] $\text{LiNiO}_2$  by which the surface is covered with lithium carbonate shows the electric discharge performance which excelled [ 60 / 60 ] even in after neglect for 20 days so that drawing 3 may show, but as for  $\text{LiNiO}_2$  with which the surface is not covered at all, electric discharge capacity has deteriorated.

[0020]The same effect is acquired, although the carbon material in which lithium carries out adsorption or an in hawk rate was used as an anode in this example, in addition even if it uses a lithium metal and a lithium alloy. Even if it transposed some nickel to other transition metals, the almost same result was obtained.

[0021]The same result was obtained even if the film of lithium carbonate was in the state which has covered only the part, without covering the whole surface of  $\text{LiNiO}_2$  completely.

[0022]

[Effect of the Invention]As mentioned above, in this invention,  $\text{LiNiO}_2$  by which the surface was covered by lithium carbonate is used as quality of cathode active material so that clearly. Therefore, the contact surface of  $\text{LiNiO}_2$  and nonaqueous electrolyte can be decreased, formation of the film which checks the electric discharge reaction an electrolysis solution is considered to be with the ability to decompose and do can be lost, and the nonaqueous electrolyte rechargeable battery which was excellent in the electric discharge performance after high temperature preservation can be obtained.

## [Brief Description of the Drawings]

[Drawing 1] The model figure explaining this invention

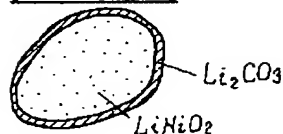
[Drawing 2] The longitudinal section of the cylindrical shape battery in the embodiment of this invention

[Drawing 3] The figure showing comparison of the electric discharge performance of this invention and a comparative example battery

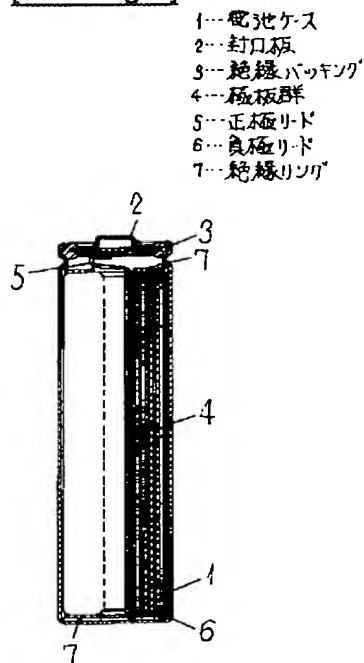
## [Explanations of letters or numerals]

- 1 Cell case
- 2 Obturation board
- 3 Insulating packing
- 4 They are plates very much.
- 5 Cathode lead
- 6 Anode lead
- 7 Insulating ring

## [Drawing 1]

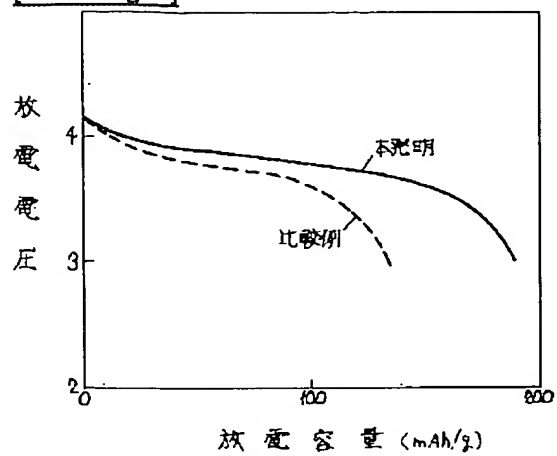


## [Drawing 2]





[Drawing 3]



---

[Translation done.]

# **Evidence Exhibit 1**

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

**Applicant:** Takanori ITOU et al.  
**Title:** POSITIVE ELECTRODE  
MATERIAL FOR NON-  
AQUEOUS ELECTROLYTE  
LITHIUM ION BATTERY AND  
BATTERY USING THE SAME



**Appl. No.:** 10/581,858  
**International Filing Date:** 11/29/2004  
**371(c) Date:** 06/30/06  
**Examiner:** Jonathan G. LEONG  
**Art Unit:** 1725  
**Confirmation Number:** 4646

**DECLARATION UNDER 37 C.F.R. § 1.132**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

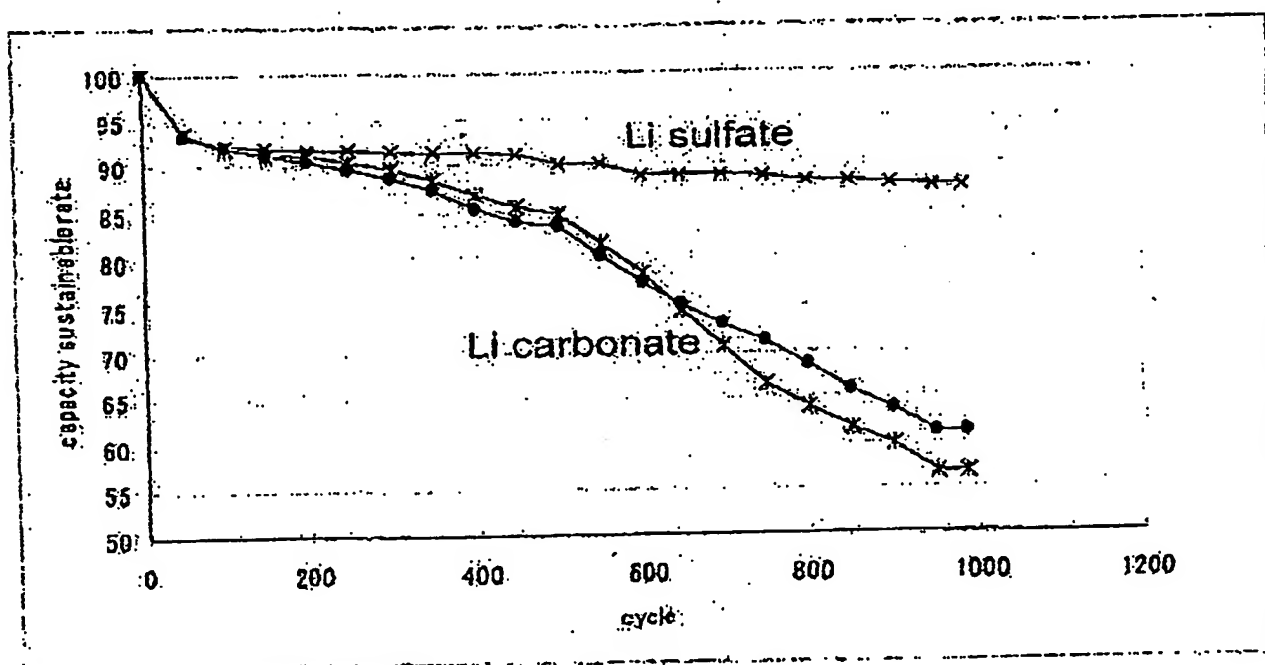
Sir:

I, Takamitsu Saito, being duly warned, hereby declare and say that:

1. I am a citizen of Japan, residing at 11-5-3-702, Shimonamiki, Kawasaki-ku, Kawasaki-shi, Kanagawa 210-0025 Japan..
2. I am now employed by Nissan Motor Co., Ltd.
3. I am a co-inventor of the invention disclosed and claimed in the above-referenced application.
4. There are two purposes for this Declaration. The first purpose is to describe the results achieved during experiments related to the claimed invention. The second purpose is

to provide comments on Japanese Publication No. 07-245105, U.S. Patent No. 5,427,875, and U.S. Patent Publication No. 2003/0157409, which have been cited against this application.

5. Discussion of Experimental Results. I performed experiments comparing the decrease in capacity of non-aqueous electrolyte lithium ion batteries with positive electrodes comprising a lithium nickel oxide, on the surface of which various different lithium compounds are deposited. Two of the lithium compounds tested were lithium sulfate and lithium carbonate. The results of the experiments are depicted in the following graph, which shows the number of cycles through which the battery was tested on the horizontal axis (up to 1000 cycles) and the percentage of initial battery capacity on the vertical axis.



6. I have extensive experience in the design of non-aqueous electrolyte batteries, and particularly in the design of lithium ion batteries. My sincere belief is that the combination of a lithium nickel oxide and a lithium sulfate being deposited is a positive electrode for a non-aqueous electrolyte lithium ion battery was never known, used, or suggested. This is why the present application was filed. In fact, the essence of the invention of claim 1 is that the combination of an oxide containing lithium and nickel and a lithium sulfate being deposited

provides dramatic improvement in the sustainable capacity of a lithium ion battery. This improvement in sustainable capacity is so dramatic as to be entirely unexpected.

7. The entirely unexpected results from this combination are shown in the above graph. Specifically, the graph shows that the battery with a lithium sulfate being deposited on the oxide of the positive electrode retained nearly 90% of its initial capacity after 1000 cycles. The battery with a lithium carbonate being deposited on the oxide of the positive electrode retained, on average, only 58% of its initial capacity after 1000 cycles. The superiority of lithium sulfate for this use was entirely unexpected.

8. Batteries using a lithium nickel oxide as the positive electrode material suffer from the problem that oxygen ions are oxidized by nickel ions with high valence within the positive electrode material into oxygen radicals and released to decompose the electrolyte solution in the battery. Lithium sulfate, when deposited onto the lithium nickel oxide, is drastically superior to lithium carbonate at preventing oxygen radicals from being emitted into the electrolyte solution. The results achieved by using lithium sulfate instead of lithium carbonate were entirely unexpected.

9. Comments on the Prior Art. As previously mentioned, I have extensive experience in the design of non-aqueous electrolyte batteries, and particularly in the design of lithium ion batteries. My sincere belief is that a person skilled in my field would not have looked U.S. Patent Publication No. 2003/0157409 ("Huang") to address the problems solved by the present invention. A person skilled in the design of lithium ion batteries would not have been motivated to combine Huang with either Japanese Publication No. 07-245105 or U.S. Patent No. 5,427,875 to create a positive electrode material comprising a lithium nickel oxide and a lithium sulfate coating. Huang simply lists a variety of possible materials out of which a slurry can be created to coat a positive electrode. Huang never suggests depositing lithium sulfate onto a positive electrode comprising lithium nickel oxide, so it could not possibly recognize the benefits of a lithium sulfate being deposited on a lithium nickel oxide positive electrode. Lithium sulfate is simply one of the twenty-two (22) possible slurry materials listed in claim 5 of Huang. In fact, lithium sulfate seems to have been added to claim 5 of Huang as an afterthought, because the specification of Huang does not even mention the

possibility of using lithium sulfate as a slurry material, much less a slurry material deposited on a lithium nickel oxide positive electrode.

10. I further declare that all statement made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent resulting therefrom.

Date: 1/24/2011

Signature: Takamitsu Saito

Takamitsu Saito